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Elementary properties of surface polaritons in a thin film

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Three quantities associated with light scattering by surface polaritons in a thin film are derived by classical electromagnetic theory. These are the excitation frequencies of these modes, the magnitude of the thermally excited electric field fluctuations and frequency dependent damping function. The excitation frequencies show that there are two branches of surface polaritons and we show that they have approximately the same linewidth at frequencies not close to the TO mode. Results are applied to a sample of InSb.

1. Introduction

This paper is an extension of the study of properties of bulk polaritons by Loudon (1970) and surface polaritons at a single interface by Nkoma *et al*(1974) to surface polaritons propagating along the interfaces of a thin film. This geometry is important because of experiments by Evans *et al* (1973) and Prieur *et al* (1975) in which surface polaritons have been observed by forward Raman scattering through a thin film. We consider a three layered structure such that a dielectric of a positive dielectric constant ϵ_1 occupies the space $z > 0$, a thin film of a frequency dependent dielectric function $\epsilon(\omega)$ occupies the space $0 < z < -d$, and the remaining space $z < -d$ is occupied by a substrate of dielectric constant $\epsilon_3 > 0$ The dispersion relation of surface polaritions in this configuration is well known to be (Mills & Maradudin 1973)

$$[\epsilon_1\alpha_2+\epsilon(\omega)\alpha_1][\epsilon_3\alpha_2+\epsilon(\omega)\alpha_3]-\exp(-2\alpha_2 d)[\epsilon_1\alpha_2-\epsilon(\omega)\alpha_1][\epsilon_3\alpha_2-\epsilon(\omega)\alpha_3] = 0 \quad \ldots(1)$$

where
$$\alpha_m{}^2 = k^2{}_{\|}-\epsilon_m \frac{\omega^2}{c^2}$$

α_m is the normal component of the surface polariton wavevectors in region $m = 1, 2, 3$. The wavevector parallel to the interfaces is denoted by $k_{\|}$ and is a conserved quantity. ϵ_m is the dielectric function in medium m, where $\epsilon_2 = \epsilon(\omega)$. Equation 1 can be solved numerically, and for a dielectric function of the form

$$\epsilon(\omega) = \epsilon_\infty+S\omega_T{}^2/(\omega_T{}^2-\omega^2) \quad \ldots \ (3)$$

typical solutions are illustrated in figure 1. In eqn 3, ϵ_∞ is the high freauency dielectric function, S measures the strength of the resonance and ω_T is the TO phonon frequency. For each $k_{\|}$ there are two frequencies corresponding to the lower mode (LM) and upper mode (UM), and agreement between theory and experiment for eq. (1) is good as shown by experiments of Evans *et al* (1973), Prieur & Ushioda (1975).

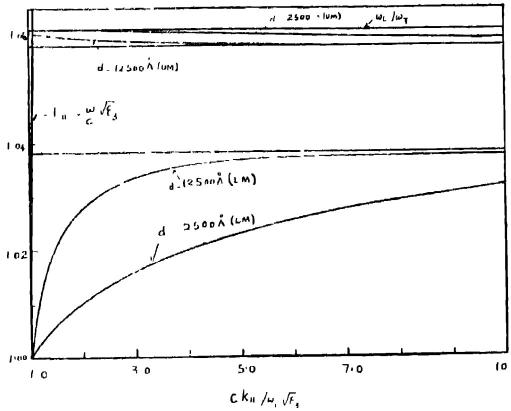

Fig. 1 Phonon-type surface polaritons dispersion curves for a thin film of InSb bounded by air ($\epsilon_1 = 1 \cdot 0$) and a substrate ($\epsilon_3 = 10 \cdot 41$) For InSb $\epsilon_\infty = 15 \cdot 7$, $S = 2 \cdot 0$, $\omega_T = 179 \cdot 0$ cm^{-1}, $\omega_L = 1 \cdot 062\ \omega_T$

In section 2 we shall consider the energetic properties, namely the total Poynting vector, energy density and demonstrate that energy velocity equals group velocity in the absence of damping. The evaluation of the energy density is a significant step, since from it one can calculate the expectation value of the squared electric field operator as in section 3.

This later quantity gives the magnitude of the fluctuations that scatter light in a Raman scattering experiment.

The energy density is also used insection 4 to calculate the frequency dependent damping function. The result agrees with that obtained by using the method of Elson and Ritchie (1972), which involves putting $\omega^2 \rightarrow \omega^2 - i\ \omega\Gamma(\omega)$ in the dispersion relation and then evaluate $\Gamma(\omega)$ to first order.

2. Energetic Considerations

2.1. *The Poynting Vector*

Surface polariton fields are solutions of Maxwell's equations in the three regions (for example see Mills & Maradudin 1973) From the fields we obtain the cycle average of the energy flow in the z-direction vanishes. The cycle average of the energy flow in the x-direction is integrated over z as

$$<I_x> = -\tfrac{1}{2}\, Re \left\{ \int_{-\infty}^{-d} E_{3z}H_{3y}{}^{*}dz + \int_{-d}^{0} E_{2z}H_{2y}{}^{*}dz + \int_{0}^{\infty} E_{1z}H_{1y}{}^{*}dz \right\} \quad \dots \ (4)$$

where E_{mz} and H_{my} are the real electric and magnetic fields one obtains from eqn. (4).

$$<I_x> = Nk_{\|}\{BC^{-}+DC^{+}+F\}\,|E_2(0)|^2 \quad \dots \ (5)$$

where
$$N = \frac{c_0\omega\epsilon(\omega)[\epsilon_1\alpha_2-\epsilon(\omega)\alpha_1]}{8\alpha_2[\epsilon^1(\omega)\alpha_1{}^2+\epsilon_1{}^2k_{\|}{}^2][\epsilon_1\alpha_2+\epsilon(\omega)\alpha_1]} \quad \dots \ (6)$$

$$B = 2\epsilon_1\epsilon_3+\epsilon^2(\omega)[(\alpha_1/\alpha_3)+(\alpha_3/\alpha_1)] \quad \dots \ (7)$$

$$C^{\pm} = 1 \pm \exp(-2\alpha_2 d)$$

$$D = \epsilon_1\epsilon(\omega)[(\alpha_2/\alpha_3)+(\alpha_3/\alpha_2)]+\epsilon_3\epsilon(\omega)[(\alpha_1/\alpha_2)+(\alpha_2/\alpha_1)] \quad \dots \ (9)$$

$$F = (2d/\alpha_2)[\epsilon_1\alpha_2+\epsilon(\omega)\alpha_1][\epsilon_3\alpha_2+\epsilon(\omega)\alpha_3] \quad \dots \ (10)$$

$|E_2(0)|^2$ is the total field in the thin film evaluated at $z = 0$.

2.2. *The Energy Density*

According to Landau and Lifshitz (1960) the energy density is

$$U_m = \tfrac{1}{2}\left\{\epsilon_0 \frac{\partial}{\partial\omega}\,[\omega\epsilon_m]\,\boldsymbol{E}_m+\mu_0\boldsymbol{H}_m\right\} \quad \dots \ (11)$$

where $\boldsymbol{E}_m$ and $\boldsymbol{H}_m$ are the real electric and magnetic fields in medium m. The total cycle averaged energy density integrated over z is

$$<U> = \int_{-\infty}^{-d} U_3dz+\int_{-d}^{0} U_2dz+\int_{0}^{\infty} U_1dz \quad \dots \ (12)$$

$$= N\,|E_2(\omega)|^2\left\{\epsilon(\omega)\,\frac{\omega}{c^2}\,[GC^{-}+JC^{+}+F]+\right.$$

$$\left.+\frac{\partial\epsilon}{\partial\omega}(\omega)\left[KC^{-}+LC^{+}+E_2\,\frac{\omega^2}{c^2}\right]\right\} \quad \dots \ (13)$$

where
$$G = 2\epsilon_1\epsilon_3+\epsilon(\omega)[\epsilon_1\alpha_3/\alpha_1+\epsilon_3\alpha_1/\alpha_3] \quad \dots \ (14)$$

$$J = \epsilon_1\epsilon_3\alpha_2[1/\alpha_1+1/\alpha_3]+\epsilon(\omega)[\epsilon_1\alpha_3+\epsilon_3\alpha_1]/\alpha_2 \quad \dots \ (15)$$

$$K = \epsilon_1\epsilon_3\omega^2/c^2-2\epsilon(\omega)\alpha_1\alpha_3 \quad \dots \ (16)$$

$$L = [(\epsilon(\omega)\omega^2/2\alpha_2c^2)-\alpha_2][\epsilon_1\alpha_3+\epsilon_3\alpha_1] \quad \dots \ (17)$$

In the absence of damping, it is easy to verify that

$$<I_x>/<U> = \frac{\partial \omega}{\partial k_{\|}} \quad \ldots \quad (18)$$

where the left hand side is the energy velocity V_E, which can be evaluated from eqns. (5) and (13) while the right hand side is the surface polariton group velocity V_G, which can be evaluated from eqn. (1). Figure 2 shows the frequency dependence of the energy velocity.

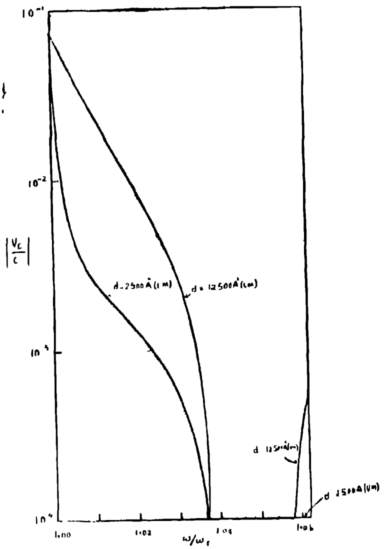

Fig 2 Frequency dependence of the phonon-type surface polariton energy velocity for the same geometry and parameters of figure 1.

3. Quantisation

The correspondence between the polariton energy given by the usual quantum harmonic oscillator expression and the classical energy density is

$$A<U> = [n_q+\tfrac{1}{2}]\hbar\omega \quad (19)$$

where A is the area of the thin film at $z = 0$, and n_q is the number of excited quanta given by the Bose-Einstein factor. In the transition from classical to quantum mechanics, the classical cycle averaged square of the electric fields as in eqn. (13)

must be replaced by the quantum mechanical expectation value of the square of the electric-field operator, which finally is obtained as

$$<n_q\,|\,E_2^2(0)\,|\,n_q> \;=\; (\hbar\omega/2A)[n_q+\tfrac{1}{2}]\gamma_q^2 \qquad \dots \quad (20)$$

where

$$\gamma_1^2 = \{<\nu>/\,|\,E_2(0)\,|^2\}^{-1} \qquad \dots \quad (21)$$

The Raman scattering cross-section by surface polaritons in a thin film is determined partly by electric field fluctuations given in eq (20) and also by fluctuations in the vibrational modes. The Raman scattering cross-section for surface polaritons has been derived by Chen *et al* (1975) and by Nkoma (1975)

4. The Damping Function

The frequency dependent damping function, Γ_w is proportional to the kinetic energy, U_{KE}, of the oscillators inside the film, and to lowest order in Γ we obtain

$$\Gamma(\omega) = \frac{2\Gamma<U_{KE}>}{<U>} \qquad \dots \quad (22)$$

$$= \frac{\Gamma\,\dfrac{\partial\epsilon(\omega)}{\partial\omega}\,\{KC^- + LC^+ + F\}}{\left\{\epsilon(\omega)\,\dfrac{\omega}{c^2}GC^- + JC^+ + F] + \dfrac{\partial\epsilon(\omega)}{\partial\omega}\left[KC^- + LC^+ + \dfrac{F}{2}\dfrac{\omega}{c^2}\right]\right\}} \qquad \dots \quad (23)$$

Figure 3 illustrates the frequecy dependence of the damping function with frequency for two film thicknesses, $d = 2500$ Å and $d = 12500$ Å

5. Conclusion

In light scattering from surface polaritons in a thin film one needs apart from the usual polarisation and symmetry consideration, first the position of the modes given by eq. 1 Secondly one needs the magnitude of the fluctuations which scatter the light, and this has been given as eq (20) obtained by quantising the surface polariton fields inside the thin film. These fluctuations can also be found by flucuation-dissipation formalism as by Nkoma (1975). Thirdly, an estimate of the magnitude of the linewidth has been obtained in section 4. In figure 2 it can be noted that at frequencies not close to ω_T, both the LM and UM have approximately the same linewidth. This has been pointed out experimentally by Prieur & Ushioda(1975). Further the form used in eq. (22) to determine the damping function has been used Ushioda & McMullen, (1972) in fitting experimental measurement of the damping function for bulk polaritons in GaP, and a similar measurement can be performed for surface polaritons. Although these experiments are difficult to perform, the study of the frequency dependence of

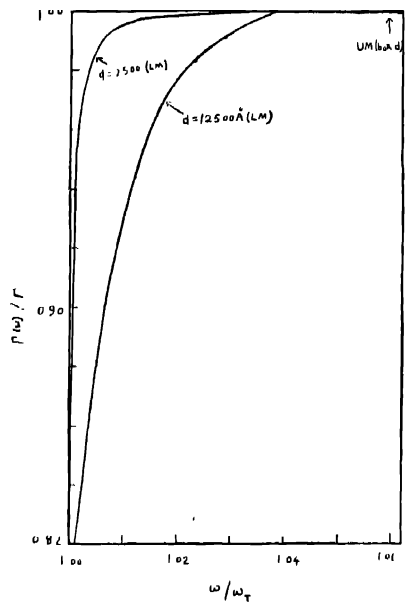

Fig. 3. Frequency dependence of the surface polariton linewidth for the same geometry and parameters as of figure 1.

the polariton linewidth offers a powerful method of obtaining the freqpency dependence of the phonon proper self energy, $\pi(\omega) = \Delta(\omega)+i\Gamma(\omega)$.

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